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● *Analyses and Modeling for Internal Dose Estimates*

ESTIMATION OF RADIONUCLIDE INGESTION: THE "PATHWAY" FOOD-CHAIN MODEL*

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Abstract—This paper describes the structure of the dynamic food-chain model PATHWAY and its utility for estimating radionuclide ingestion after fallout deposition from nuclear testing in Nevada. Model input requirements are described and output examples are provided. The basic output of PATHWAY is the time-integrated radionuclide ingestion by humans per unit fallout deposition (Bq per Bq m^{-2}). Output specific to sex, age, life-style (diet), location (agricultural practice), event (calendar date), and radionuclide may be generated. Uncertainties of model predictions, based on "Monte Carlo" simulations using parameter value distributions, are described. Results of a sensitivity analysis, based on a ranking of partial correlation coefficients, are reviewed to illustrate the relative importance of parameters and associated transport pathways. Output data for ^{131}I and ^{137}Cs in milk are compared with predictions from several well known food-chain models. Preliminary efforts to validate PATHWAY results with real data sets are described.

INTRODUCTION

THE PATHWAY food-chain model was developed to predict radionuclide ingestion by residents of portions of nine western states following radioactive fallout deposition. The fallout resulted from nuclear testing at the Nevada Test Site (NTS) between 1951 and 1962 (Carter and Moghissi 1977; Friesen 1985a). Such predictions have been used to estimate internal dose by the Off-Site Radiation Exposure Review Project (ORERP), initiated in 1979 by the Nevada Operations Office of the U.S. Department of Energy (Friesen 1985b). The ORERP was established to employ the best possible methods and all available data to provide retrospective dose estimates from all internal and external pathways to the people most exposed to NTS fallout. The effort was prompted by a paper on childhood leukemia by Lyon et al. (1979), public and congressional concern (Subcommittee on Oversight and Investigations 1980), and the case of Allen vs. United States (Civil Action No. C-79-515, U.S. District Court for the District of Utah Central Division, filed 30 August 1979).

Food-chain transport models were available at the outset of the ORERP (e.g., Hoffman et al. 1978, 1984; Booth et al. 1971; Baker et al. 1976; U.S. Nuclear Regulatory Commission 1977); however, none were found that met all requirements of this effort. PATHWAY uses many concepts and parameters embodied in earlier models (e.g., those cited above plus Pleasant et al. 1980; Simmonds and Linsley 1981), but adds new features and capabilities. An incomplete description of PATHWAY was published earlier (Kirchner et al. 1983) and a much more detailed description of the model structure and parameter values was recently published (Whicker and Kirchner 1987). The intent of this paper is to provide a general overview of the model, including its utility for making radionuclide ingestion estimates and its general properties (e.g., uncertainty, parameter sensitivity and predictive accuracy).

DESCRIPTION AND STRUCTURE OF PATHWAY

PATHWAY simulates the transport of some 21 fallout radionuclides (^{131,133,135}I, ^{136,137}Cs, ^{89,90,91}Sr, ^{103,106}Ru, ^{143,144}Ce, ^{97}Zr, ^{99}Mo, ^{140}Ba, ^{132}Te, ^{105}Rh, ^{147}Nd, ^{239}Np, ^{93}Y, ^{239}Pu) through agricultural ecosystems to humans. The model is implemented as a FORTRAN V code and employs PREMOD and MODAID software (Kirchner and Vevea 1983). The model simultaneously simulates the flow of radionuclides through pasture, hay, rangeland and human-food crop ecosystems. Model parameters were chosen to simulate agricultural conditions of the Southwestern U.S. during the 1950s.

Each agroecosystem is represented by six compartments or state variables (Fig. 1). Each state variable represents the quantity of radionuclide (Bq m^{-2}) in a specific compartment, such as vegetation surfaces, internal tissues of vegetation, soil surface (0–0.1 cm), labile soil (0.1–25 cm), fixed soil (0.1–25 cm) and deep soil (>25 cm). In

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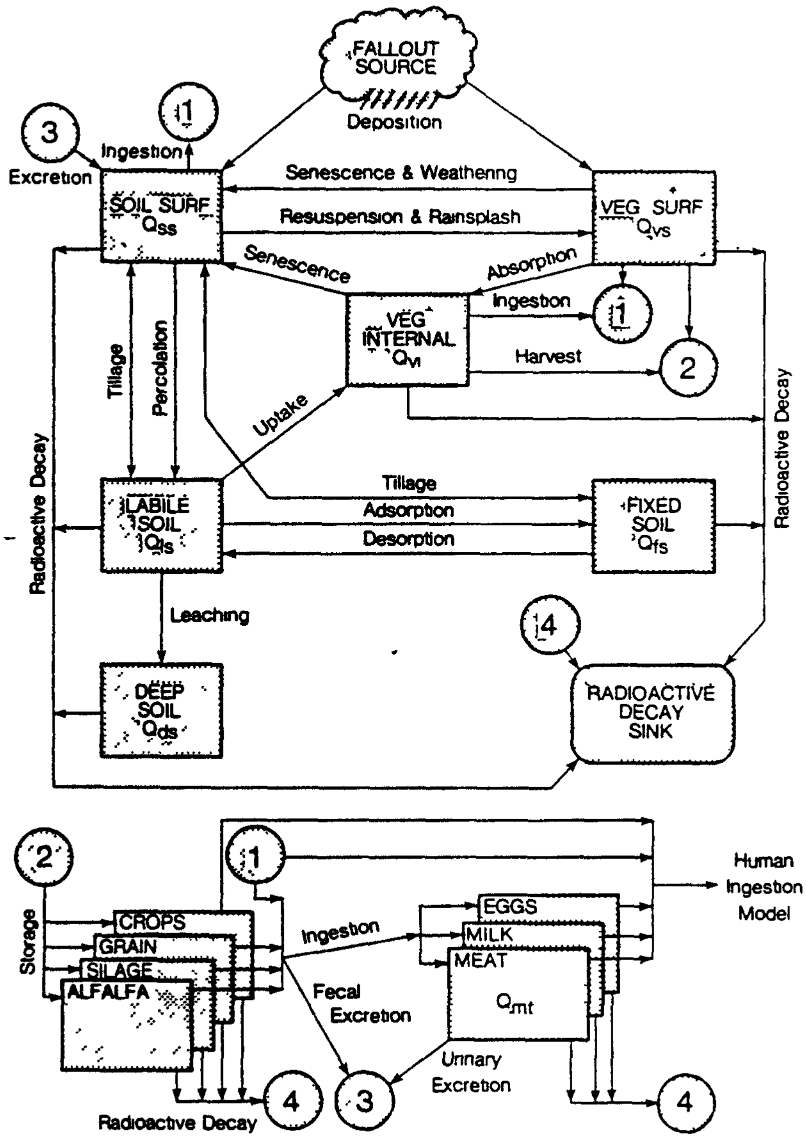

Fig 1 Structural features of the PATHWAY model Boxes represent compartments or state variables arrows represent transfers resulting from indicated processes circles connect process arrows between the upper and lower diagrams

addition, these systems contain specific compartments representing quantities or concentrations of radionuclides in fruits vegetables (leafy and other), grains grain plants for silage grass and alfalfa hay (two to six distinct annual cuttings), meats (beef lamb, and poultry), milk milk products (hard cheese, cottage cheese, and ice cream) and eggs

Discrete events and continuous processes are simulated Discrete events specified by calendar date include fallout deposition, soil tillage, crop harvest, and livestock diet changes Continuous processes include resuspension and rainsplash of surface soil to plant surfaces, weathering and senescence from plants to soil, percolation and leaching down through the soil profile, adsorption and desorption between fixed and labile soil components, root uptake from soil to plant tissues absorption of surficial material by plant tissues ingestion and excretion by animals and radioactive decay (Fig 1)

Continuous rate processes are embodied in ordinary first-order differential equations that equate the time derivatives of the state variable quantities to the summation of all inflow rates minus the summation of all outflow rates (Whicker and Kirchner 1987) A single differential equation is written for each state variable then the resulting set of coupled differential equations is solved numerically on daily time steps using a Runge-Kutta algorithm of order four (Carnahan et al 1969) The solutions provide daily inventories (Bq m^{-2}) for each compartment which may be converted to units of concentration (Bq kg^{-1}) for subsequent calculations Daily concentrations of radionuclides in human-food products such as meat milk eggs and vegetables are numerically integrated over time to provide estimates of integrated concentrations per unit fallout deposition (Bq d kg^{-1} per Bq m^{-2}) Integration times are seven half-lives for the shorter-lived radionuclides and 4 2 y for long-lived nuclides which account for >95% of the infinite time values

After modifications for washing and radioactive decay losses time-integrated concentrations in foodstuffs are multiplied by the age-, sex- and life-style (diet)-specific intake rates (kg d^{-1}) of the respective foods to obtain the integrated radionuclide intakes per unit fallout deposition (Bq per Bq m^{-2}) for each food type Summation of these values over all food types yields the total intake of a given radionuclide per unit fallout deposition Intake corrections that account for consumption of imported or stored foods that would not be contaminated are made The intake per unit deposition when multiplied by the measured or estimated fallout deposition (Bq m^{-2}) and by the appropriate organ dose conversion factor (Gy Bq^{-1}) yields an organ-specific dose estimate for a particular radionuclide The same procedure is repeated for each radionuclide of interest so that the radionuclide-specific organ doses may be summed to provide a total internal organ dose estimate for all radionuclides combined (Whicker and Kirchner 1987)

Several special features and capabilities are incorporated in PATHWAY For example the growth senescence and harvest of plants is simulated through the calendar year as well as the storage of hay for future consumption by livestock (Whicker and Kirchner 1987) Livestock diets are specified combinations of pasture hay, grain and silage these combinations vary through seasons (Fig 2) Diet variability has a dramatic effect on the concentrations of radionuclides in food products Various agricultural regimes can be incorporated into the model Important factors in this regard include in addition to the livestock diets mentioned above milk distribution patterns pasture seasons geographic sources of animal feeds hay harvest dates and food product storage times

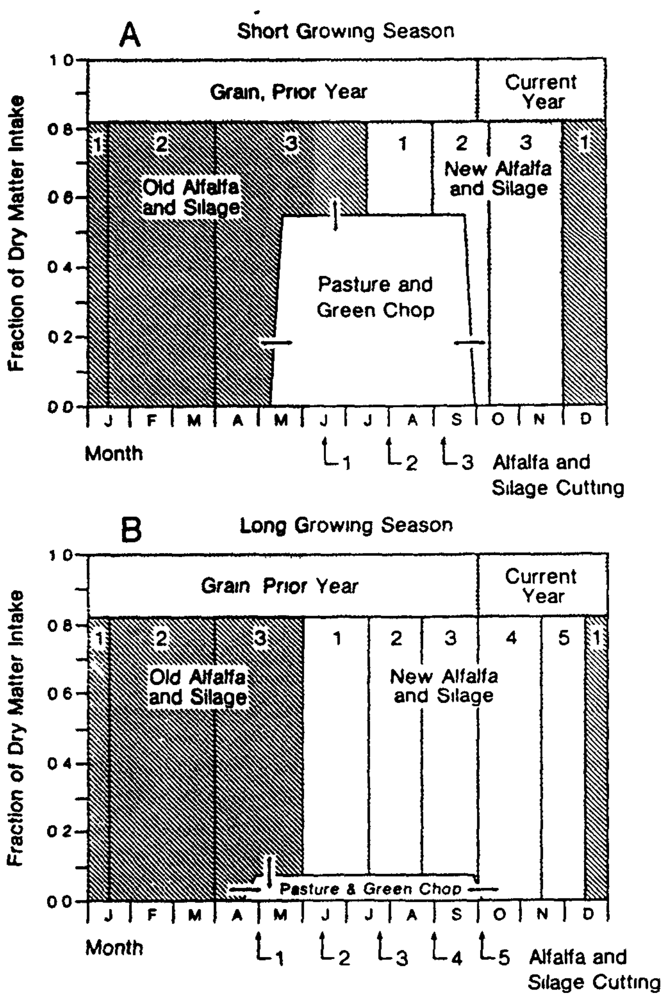

Fig 2 Dairy cow diets assumed for A short growing season areas (upper diagram) and B, long growing season areas (lower diagram) in the semi-arid Western U S

3

A data base on these factors covering portions of a nine-state area was compiled for the 1950s period (Ward and Whicker 1987) A life-style survey of 10 southwestern counties near the NTS was also conducted to provide region-specific information on dietary and food production practices of the rural and small communities for the same time PATHWAY can be run in either deterministic (single parameter values) or stochastic (random selection from parameter distributions) modes The stochastic mode provides estimates of model output uncertainty, as well as information enabling a parameter sensitivity analysis that reveals the relative importance of uncertain parameters on the model output (Otis 1983)

MODEL INPUT REQUIREMENTS FOR AVAILABLE OUTPUT OPTIONS

The capabilities and flexibility of the PATHWAY code impose numerous input requirements The requirements vary, depending on purpose of the run and the forms of output desired

Fallout deposition

In many cases, the desired model output is the integrated radionuclide intake per unit fallout deposition For such cases, it is not necessary to know the radionuclide deposition If, however, a large geographic area is contaminated with fallout, but to differing degrees, and human or livestock food produced within different portions of the area is shared or pooled in some manner, it is necessary to input several radionuclide-specific deposition estimates into PATHWAY For example in constructing intake estimates for ^{131}I via milk, it was found that NTS fallout patterns were nonuniform within milk sheds Since milk entering most major processing centers was produced from different geographic areas, it was necessary to determine fallout deposition in each milk producing area It was also necessary to estimate the relative contribution of each milk source to the milk processor and the contributions of various processors (or individual dairies or cows) to the intake of any particular individual Furthermore, it was necessary to estimate the critical agricultural parameters (e g , hay sources and harvest times pasture season and degree of pasture consumption) for each milk source Such networking capability has been achieved with PATHWAY, but the input data requirements are substantial (Ward and Whicker 1987)

The time-dependency of PATHWAY leads to radionuclide intakes per unit deposition that vary by calendar date of the fallout event These variations are dramatic for short-lived radionuclides (Whicker and Kirchner 1987) Therefore specification of the calendar date(s) of fallout deposition is essential PATHWAY is normally employed to deal with acute deposition events, however, it is readily capable of making human intake or food concentration estimates for continuous deposition scenarios In the latter case, the input requirement is an estimate of the daily deposition of a specific radionuclide The daily deposition could either be constant or time-dependent An indication of fallout particle size is important because the initial partitioning of deposition between the soil and vegetation is dependent on the foliar interception constant which appears to decrease with particle size giving rise to a smaller foliar deposition fraction[§] (Whicker and Kirchner 1987, Anspaugh et al 1986 Romney et al 1963)

Model parameters

PATHWAY embodies numerous parameters some of which are radionuclide-dependent At present, data for 21 radionuclides representing 14 elements are contained in the model's files Other isotopes of these elements may be easily handled by simply entering the appropriate radioactive decay constant If other elements are to be modeled, it is necessary to provide data on parameters such as the plant/soil concentration ratio, assimilation fractions to meats, milk and eggs, elimination rate constants for meats, soil adsorption-desorption rate constants soil leaching rate constants, and foliar absorption rate constants (Whicker and Kirchner 1987)

Many parameters in the model are independent of the radionuclide in question These generally describe physical and biological transport processes and activities controlled by man (e g , agricultural practices, human diet food preparation, food distribution patterns, etc) These parameters may vary significantly by season, geographic location, current weather, plant and soil characteristics and other factors Values for all such parameters have been estimated with specific reference, where possible to the agricultural conditions prevailing during the 1950s in the arid to semi-arid western U S Agricultural practices data have been compiled on a community- or county-level basis for portions of nine western states (Ward and Whicker 1987) and such data are contained in the files of PATHWAY Data for counties are specified for rural (<2,000 residents), town (2,000–25 000), and urban (>25,000) categories

Target individuals or populations

The model is readily adaptable to internal dose estimation for specified or unspecified individuals within particular population groups In the case of specified individuals, it may be possible to incorporate specific information on residence history and food consumption patterns (e g , sources types and amounts of foods consumed) For the case of Allen vs United States some 24 litigants (or their representatives) were individually invited to provide specific data "Default" parameter value estimates were used in cases where the litigants were unable to provide a specific estimate For unspecified individuals "default" parameter estimates on file are utilized

Age- and sex-specific data on food consumption rates were available These values were obtained from a 10-

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[§] Personal communication (1987) S L Simon Division of Epidemiology University of Utah Salt Lake City, UT 84103

Table 1 Arithmetic mean food consumption rates estimated from the 10-county life-style survey data Estimates are relevant to rural areas and towns (<25 000 residents) during the 1950s in the arid and semi-arid Western U S

Food Item	Daily Intake (kg) (Fresh Weight) Age 12 18 y Male	Female	Age > 19 y Male	Female
Milk	0 644	0 434	0 477	0 436
Cottage Cheese	0 033	0 065	0 070	0 073
Hard Cheese & Ice Cream	0 070	0 058	0 090	0 073
Beef[a]	0 151	0 117	0 178	0 116
Lamb	0 017	0 008	0 029	0 016
Poultry	0 041	0 035	0 049	0 041
Eggs	0 064	0 051	0 070	0 048
Leafy Vegetables	0 026	0 024	0 030	0 029
Other Vegetables & Fruits	0 290	0 253	0 282	0 269
Grains	0 084	0 061	0 090	0 066

[a] Includes beef, pork and venison consumption

county life-style survey[‖] and from summaries prepared by Rupp (1980) The life-style survey data have been used for teens and adults living in rural areas and towns with <25,000 residents, corresponding to the population strata sampled (Table 1) The Rupp (1980) data summaries have been employed for teens and adults in urban (>25,000 people) locations and for infants and children, irregardless of community size (Whicker and Kirchner 1987)

Deterministic vs stochastic mode

When single-value output estimates are desired, the model is run in the deterministic mode Parameters are usually taken as the "most probable" or "best estimate" values although arithmetic mean median, or other values could be used The default single-value estimates for all model parameters are found in Whicker and Kirchner (1987)

If estimates of output uncertainty are desired, PATHWAY can be run in the stochastic mode This imposes the requirement that for all parameters deemed uncertain, the distribution of all reasonably possible values be specified The form of the distribution (e g., normal, normal-truncated, lognormal triangular, uniform, etc) must be specified as well as the distributional statistics (e g , mean and standard deviation, geometric mean and standard deviation, mode, upper and lower limits, etc) Distributions should be selected to minimize bias (Rose 1983) and eliminate unrealistic simulations (O'Neill et al 1982) When PATHWAY is operated in the stochastic mode, a random or "Monte Carlo" sampling of all uncertain parameter values precedes each model run

Any number of runs may be specified Ordinarily, 100–1000 runs provides a stable estimate of the output distribution Various statistical parameters describing the output data are provided by PATHWAY (e g , geometric means and standard deviations, arithmetic means and standard deviations, skewness and kurtosis of the log-transformed and untransformed data minimum and maximum values, etc) Frequency histograms as well as cumulative frequency plots can also be provided (Otis 1983)

A parameter sensitivity analysis may be performed on stochastic input/output data The selected parameter values are treated as independent variables and the resulting, respective output values as the dependent variables A multiple regression procedure is employed to calculate partial correlation coefficients for each of the independent variables (Otis 1983, Breshears 1987) These may be ranked to indicate the relative degree of influence of each parameter on the model output assuming parameters are independent This procedure is particularly useful because the influence of each uncertain parameter may be tested under the condition where all other uncertain parameters may vary over their specified range (Otis 1983)

Model validation

Validation exercises to test predictive accuracy impose various requirements on the model input depending

[‖] The 10-county survey was conducted in Utah (Kane Washington and Iron Counties) Nevada (Lincoln White Pine Nye and Esmeralda Counties) and Arizona (Coconino and Mohave Counties) on the form of the real data to which the output will be compared Most of the model validation exercises completed to date for PATHWAY have involved real data sets on radionuclide concentrations in milk meat or forages (Kirchner and Whicker 1984) Such data sets are particularly useful when time-series values are available as well as information on local agricultural practices and conditions However, such data are not particularly useful in testing PATHWAY unless corresponding data on local fallout deposition are available or can be estimated When such data are available, the fallout deposition data can be entered into the model, parameters can be adjusted as necessary to assure compatibility with prevailing local conditions, and the predicted radionuclide concentrations in the various food products can be generated as a function of time

The stochastic mode can be used to estimate the predictive uncertainty This is useful, since the geometric means of the stochastic output distributions are very close (numerically) to the deterministic estimates, and the magnitude of predictive uncertainty should be considered in validation work Model output can then be compared using various statistical procedures, with the real data set (Kirchner and Whicker 1984)

Other forms of model output are sometimes useful, and PATHWAY can provide flexible output options For example, cumulative inventories of long-lived radionuclides in the soil profile can be estimated over long time periods if time-dependent fallout deposition estimates are available Time-integrated concentrations following acute deposition events can be provided by the model for comparison to similar real data, or for comparison to output of steady-state models that estimate equilibrium food concentrations per unit of chronic deposition rate (Subcommittee on Risk Assessment for Radionuclides 1984) Finally, PATHWAY can provide, for example, ratios of variables (e g , milk/vegetation) to compare with such real data for testing a smaller segment of the food-chain process

UNCERTAINTY OF MODEL OUTPUT

Stochastic runs to estimate model output uncertainty were conducted for ^{131}I, ^{136}Cs and ^{137}Cs in milk meats, eggs, vegetables and animal forage (Otis 1983) Iodine-131 and ^{137}Cs represent potentially important dose contributors The choice of ^{136}Cs permits comparison of a short- vs long-lived isotope of a common element Six different calendar dates, representing different NTS fallout events, were chosen for study to determine whether timing of a fallout event had a major effect on uncertainty estimates Forms of output studied included time-dependent and time-integrated radionuclide concentrations in the various food items Stochastic parameters for this initial work included the rainsplash and weathering rate constants, the resuspension factor, the foliar interception constant, assimilation fractions to milk, meat, or eggs, the dry matter ingestion rate of livestock, the biological elimination rate constant, the production rate of milk or eggs, the foliar absorption rate constant and the plant/soil concentration factor (Otis 1983 Whicker and Kirchner 1987) All other parameters including those describing pasture use by dairy cows were treated as constants

A general result of the uncertainty analyses conducted by Otis (1983) was that the lognormal distribution provided a reasonable description of the output data thus the geometric standard deviation was used as the most appropriate measure of dispersion Geometric standard deviations (GSDs) of integrated concentrations were not strongly dependent on the date of fallout deposition (within the interval 17 March–30 August) and usually ranged from 1 4 to 2 0 for the three radionuclides and several foodstuffs considered Except for differences that appear to be due to half-life the GSDs did not vary greatly between radionuclides This result seems reasonable because uncertainty was caused primarily by radionuclide-independent processes Integrated concentrations of ^{137}Cs tended to have slightly higher GSDs than ^{136}Cs suggesting that a longer half-life provides more time for very uncertain processes such as resuspension to operate

Uncertainty in integrated concentrations in foods tended to be greatest soon after the fallout event after which the GSDs declined with integration time However stable values were reached well before the normal integration times Annual average GSD values for milk were 1 8 for ^{131}I and ^{136}Cs, and 1 9 for ^{137}Cs Values for vegetables were 1 7 for ^{131}I and ^{136}Cs, and 2 0 for ^{137}Cs GSDs for ^{137}Cs in beef and eggs were 1 7 and 1 9, respectively

Recently, Breshears (1987) and Breshears et al (1989) incorporated uncertainty in the dairy cow diet in the analysis in addition to the uncertainty in parameters studied by Otis (1983) Uncertainty distributions for the timing of pasture use (dates cows were placed on and off pasture) and the amount of pasture use (percent of total dry matter intake) were compiled from studies recently completed in several western states (Ward and Whicker 1987, Walters et al 1985) Two general scenarios were developed a shorter pasture season with higher average pasture use, characteristic of higher elevation areas with greater water availability, and a longer pasture season with lower average pasture use, characteristic of more desert-like areas (Fig 2) The short pasture season corresponds to the scheme of three annual harvests for alfalfa while the long season corresponds to the scheme of five annual harvests Statistics for pasture use parameter distributions are provided in Table 2

The additional variability in pasture use and different updated diet scenarios produced a rather pronounced effect of the date of fallout deposition on the GSD of integrated ^{131}I concentrations in milk (Breshears 1987, Breshears et al 1989) During winter (November through February), when only stored feeds were utilized the GSD values for both scenarios were only about 1 3 During the pasture season the long-season, low-pasture-use scenario produced rather constant GSDs of about 1 6 from May through September The short-season, high-pasture-use scenario produced GSDs of about 1 8 during the middle of the pasture season similar to the original values found

6

Table 2 Distributional statistics for pasture-use parameters used in an uncertainty analysis of PATHWAY Distribution types are indicated Dates are presented in Julian format followed by calendar format in parentheses

		Short Growing Season	Long Growing Season
Fraction of dry matter, intake obtained from pasture (truncated normal)	mean	0 55	0 079
	s d	0 13	0 075
	lower bound	0 00	0.00
	upper bound	0 82	0.82
Date cows were placed on pasture (triangular)	minimum	115 (Apr 25)	98 (Apr 8)
	mode	135 (May 15)	116 (Apr 26)
	maximum	159 (Jun 8)	142 (May 22)
Date cows were taken off pasture (triangular)	minimum	236 (Aug 24)	251 (Sep 8)
	mode	265 (Sep 22)	276 (Oct 3)
	maximum	280 (Oct 7)	295 (Oct 22)

by Otis (1983) However, during the pasture transition periods the uncertainty in dates cows were going on or off pasture produced GSDs approaching 2 1 in late spring and 2 4 at the end of the summer

This finding indicates that for the high-pasture-use scenario uncertainty in the timing of pasture use produces considerable increases in the uncertainty of integrated ^{131}I concentrations in milk in May and September Similar stochastic runs for ^{137}Cs in milk indicated that pasture use uncertainty added little uncertainty to the model output Values of the GSD for ^{137}Cs in milk ranged from 1 4 to 1 8, all less than the value of 1 9 obtained by Otis (1983)

Distributional data on food consumption rates of people resulting from the 10-county life-style survey made possible stochastic runs in which the uncertainty of the total integrated intakes per unit deposition could be evaluated Runs for ^{131}I and ^{137}Cs intakes from all food sources were evaluated The actual, empirical distributions of food intakes were used because in many cases, these were not well-described by a particular function It was found however that the resulting distributions for the output were usually reasonably approximated by the lognormal form

GSDs for integrated intakes from all sources generally ranged from 2 0–2 4 Within a food type (e g , milk) uncertainty in the consumption rate produced an increase from a GSD of 1 8 in integrated concentration to a value of 2 4 for integrated intake However when all food sources were summed, lower GSDs resulted, with values generally ranging from 2 0 to 2 2

SENSITIVITY OF MODEL OUTPUT TO INDIVIDUAL PARAMETERS

Multiple regressions on results from the uncertainty analyses provided partial correlation coefficients for each stochastic parameter (Otis 1983, Breshears 1987) Partial correlation coefficients (PCCs) provide a measure of the sensitivity of model output to variable input parameters (Gardner et al 1980), assuming parameters vary independently (Rose 1983) The method we used for sensitivity analysis was "global," in that all parameters were permitted to vary over the entire range of values judged possible Parameters can affect the model output both by virtue of their degree of functional importance in the model, and by the magnitude of uncertainty in their values We did not distinguish between these types of effects

Perhaps the most striking result of the parameter sensitivity analyses was the dominance of plant contamination processes in affecting the integrated concentrations of ^{131}I ^{136}Cs, and ^{137}Cs in food products The dominance of parameters affecting such processes was indicated by the frequent occurrence of PCCs in excess of about 0 7 For ^{131}I and ^{136}Cs, the foliar interception constant which determines the fraction of a fallout deposit that is initially intercepted by foliage, consistently had the highest ranking PCC This parameter was usually followed by the resuspension factor in the case of meat and vegetables and by the cow s milk production rate in the case of milk

The resuspension factor was most influential for ^{137}Cs in food products with the foliar interception constant usually ranking second or third The difference in parameter rankings between the short- and long-lived radionuclides may be explained on the grounds that resuspension can become dominant for long-lived materials by having more time to operate The effect of the foliar interception constant was most strongly manifest near the beginning of a simulation (Otis 1983) The PCCs for the foliar interception constant dominated for the first 20 d thereafter the resuspension factor dominated The weathering rate constant, which accounts for the loss of radioactivity from foliage, had PCCs in excess of 0 7 for ^{131}I and ^{136}Cs in vegetables A pronounced effect of season during which fallout occurs on the PCCs for ^{131}I in milk was observed (Breshears 1987) Resuspension was dominant for fallout events in winter but the foliar interception constant dominated during the pasture season (Otis 1983)

A few parameters were frequently observed to have PCCs of 0.3 to 0.7, which revealed their importance but not dominance. These included the weathering rate and rainsplash constants. The assimilation fractions to meat or milk also fell in this intermediate importance category, as did the muscle elimination rate constant for meat products.

One parameter allowed to vary for the Cs isotopes, the plant/soil concentration factor, never had a significant PCC. This is indicative of the comparative unimportance of the plant root uptake process for the conditions modeled by PATHWAY. In earlier studies, the concentration factor was changed from its nominal value to zero in the case of several radionuclides, including ^{131}I and ^{90}Sr, and the output values were virtually unchanged. For this reason, less effort has been spent on root uptake than other processes. This, of course, does not rule out the possible importance of root uptake in other circumstances. Some of our validation work suggests that PATHWAY may be underestimating ^{90}Sr in the food chain several years after fallout deposition (Kirchner and Whicker 1984). It is possible that this is due to an underestimate of root uptake.

Several needs have been identified through parameter sensitivity analysis. One is the need to better define some of the more critical parameters, such as the foliar interception constant and the resuspension factor. The foliar interception constant and its uncertainty is not founded on a large data base. We believe it is related to fallout particle size and perhaps other factors, such as climatic variables and vegetation characteristics. The formulation for resuspension is grossly oversimplified for such a complex process. This is one reason why uncertainty in the resuspension factor is so large.

Another aspect that needs attention is the effect of parameter covariance. Work to date assumed that all parameters vary independently. If parameter covariances were known, the uncertainty and sensitivity analyses could be repeated, possibly with different results.

COMPARISON OF PREDICTIONS: PATHWAY VS OTHER CODES

Several internationally recognized models for assessing radionuclide transfer through terrestrial food chains were compared by Hoffman et al. (1983, 1984). Comparisons were made of the model predictions of steady-state concentrations of ^{137}Cs, ^{90}Sr and ^{131}I in milk, meat, and vegetables per unit deposition rate (Bq kg^{-1} per Bq m^{-2} d^{-1}). Models compared included AIRDOS-EPA (U.S. Environmental Protection Agency), IAEA (International Atomic Energy Agency), NRPB (National Radiological Protection Board of the United Kingdom), BIOPATH (Studsvik Energiteknick AB, Sweden), and NRC (U.S. Nuclear Regulatory Commission). In some cases, observed data compiled by UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) were compared to model predictions.

The steady-state output of these models may be compared directly to the principle output of PATHWAY because the steady-state concentration per unit deposition rate is equivalent to the infinite time-integrated concentration per unit of acute deposition, provided the same fundamental units are used. Comparisons between the nominal output values of PATHWAY and those of models compared by Hoffman et al. (1984) are shown in Figs. 3 and 4. The most striking difference between models is the strong seasonal dependence in the PATHWAY results. The PATHWAY values represent integrated concentrations in foods for an acute fallout event deposited on the dates indicated. If a chronic, constant deposition were input to the PATHWAY code, the seasonal dependencies would be less dramatic but still present.

For ^{131}I in milk (Fig. 3), the values predicted by PATHWAY are much lower than those from all other models. However, in PATHWAY, the foliar interception constant was lower than the values used in the other codes because it was intended to simulate the large fallout particles that dominate within 400 km from the NTS (Whicker and Kirchner 1987). If comparable values of α were used in PATHWAY, the ^{131}I values in milk would be about five times higher during the mid-summer pasture season. The temporal pattern of PATHWAY reflects the intake of pasture from May through September. In winter the primary source of ^{131}I intake is the ingestion of contaminated soil and dust. Hay, silage, and grain are generally stored long enough for ^{131}I to decay prior to consumption.

Comparisons for ^{137}Cs in milk are shown in Fig. 4. Maximum values for PATHWAY occur in late spring to early summer, while minimum values occur in fall and winter. Maximum values reflect the consumption of fresh pasture and hay harvested in early summer. Minimum values occur when dairy cows are consuming stored feed that would be relatively uncontaminated. Much of the fallout deposited through the fall and winter will be weathered into the soil by the time cows begin to consume

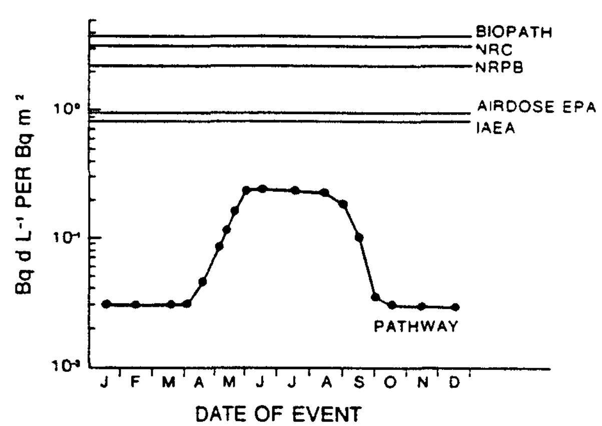

Fig. 3. Comparison of several food-chain models in the predicted time-integrated ^{131}I concentrations in milk per unit deposition vs. calendar date of the deposition event.

8

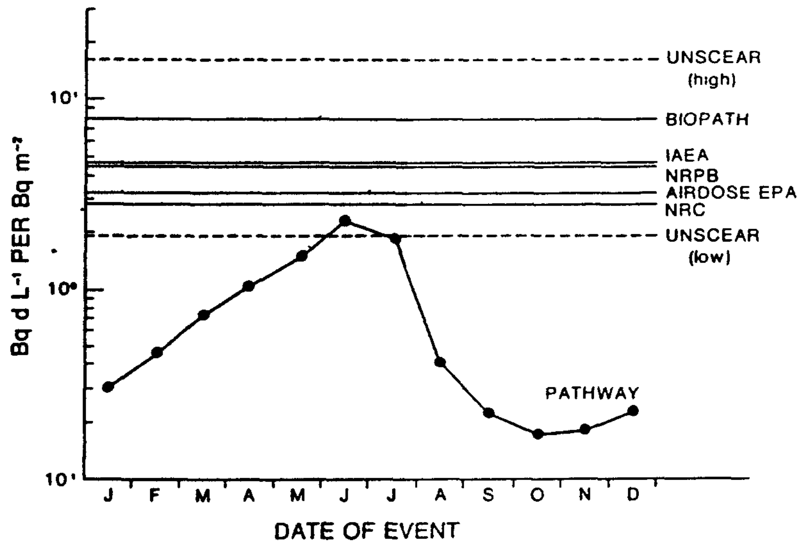

Fig. 4 Comparison of several food-chain models in the predicted time-integrated ^{137}Cs concentrations in milk per unit deposition vs calendar date of the deposition event

fresh forage in spring Cesium transfer from soil to vegetation is generally minimal in most geographic areas

Time-integrated concentrations of ^{137}Cs in range beef and lamb per unit deposition, estimated by PATHWAY, were not as strongly dependent on seasonal timing of the fallout event (Whicker and Kirchner 1987) In the case of ^{137}Cs in beef, PATHWAY estimates ranged from 4 to 9 Bq d kg^{-1} per Bq m^{-2} Minimum values occurred in early summer, and maximum values were found in fall and winter This pattern reflects the rapid growth of forage plants in early summer and "dilution" of radioactivity by new biomass Time-integrated ^{137}Cs concentrations per unit fallout deposition in pasture as estimated by PATHWAY, were minimal in May–June (20 Bq d kg^{-1} per Bq m^{-2}) and maximal in fall and winter (60 Bq d kg^{-1} per Bq m^{-2}) Values for ^{137}Cs in beef predicted by PATHWAY compared favorably to those predicted by IAEA and AIRDOS-EPA (7–8 Bq kg^{-1} per Bq m^{-2} d^{-1}) PATHWAY values for ^{137}Cs in pasture were also within a factor of two of the respective predictions by IAEA AIRDOS-EPA and NRPB (Hoffman et al 1984)

The order-of-magnitude agreement between PATHWAY and other food-chain models is not surprising because much of the same experimental data was used for parameter estimates in all models compared It is also not surprising that the models do not show better overall agreement because mathematical formulations transport processes considered, actual parameter values and model purposes varied among the codes compared Considering the magnitude of uncertainty to be expected in the output of complex food-chain models, as well as in real world systems, the differences among predictions do not, in general, seem unduly large The most significant difference between PATHWAY and the other models considered is in the degree of time-dependence

VALIDATION OF PATHWAY PREDICTION VS OBSERVATION

The degree of credibility assigned to complex food-chain model predictions should be related to the extent of testing of the model's performance A considerable effort has been made to subject PATHWAY to critical peer review and to test its predictive accuracy and dynamic properties Preliminary work on validation of the model was described previously (Kirchner and Whicker 1984) This initial work involved examination of model predictions against 37 independent data sets Each of these data sets represented observations through time of the concentrations of a radionuclide in milk, beef pasture or alfalfa Most data represented concentrations of ^{131}I ^{137}Cs and ^{90}Sr, although some data on ^{140}Ba in milk were also examined Several data sets were collected after regional fallout from single events (e g , the Smallboy test event at the NTS and subsequent measurements of ^{131}I ^{137}Cs and ^{140}Ba in milk at several locations in Nevada and Utah and the Windscale accident in England and subsequent measurements of ^{131}I in milk) Other data included measurements of ^{137}Cs and ^{90}Sr in foods over periods up to 10 y (1957–1967) in response to global fallout

Our general approach to model validation is shown for ^{131}I in milk following the Smallboy detonation at the NTS on 14 July 1962 Figure 5 illustrates ^{131}I concentrations measured by the U S Public Health Service in milk from Kamas, Oakley and, Kimball Junction UT over a period of 30 d following the Smallboy event The solid line represents the predicted values from PATHWAY based on deposition estimates from survey meter data (Knapp 1963) and conversion of exposure rates to ground deposition (Hicks 1982) Although the agreement between observations and predictions appears very good in

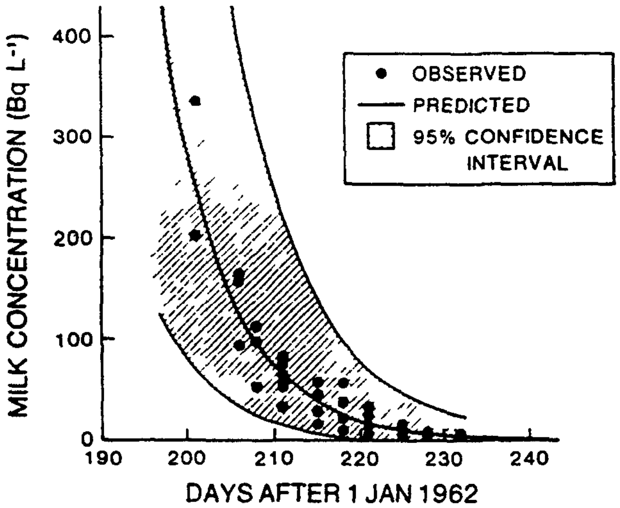

Fig 5 Observed and predicted ^{131}I concentrations in milk from Kamas, Oakley and Kimball Junction UT, following the Smallboy detonation at the Nevada Test Site on 14 July 1962

this case we chose to use statistical approaches to more objectively compare observations and predictions

First, observations are plotted against predictions after pairing the data by date (Fig 6) A correlation analysis can be applied to the data to test the hypothesis that the time-ordered pairs of data and model predictions are correlated In cases where the correlation coefficient is significant, a slope different from 1 0 indicates differences according to a scaling (multiplicative) factor, whereas a nonzero intercept indicates differences by an additive factor In the case shown the correlation coefficient was significant (r = 0 97) It may seem logical to test the hypothesis that the slope is 1 0 and the intercept is zero Unfortunately, autocorrelation of such time-series data may bias inferences about the slope and intercept (Aigner 1972), thus these tests are not statistically valid

Examination of the residuals from the regression can be used to determine whether the model dynamics represent the temporal variations of field data Runs tests are computed for each data set of residuals to identify any significant trends (Draper and Smith 1966) In the example shown there were no significant 'runs' of positive or negative residual values, indicating that the model dynamics represent the observed temporal trend

The shaded area about the line of predicted concentrations in Fig 5 represents our best estimate of uncertainty (95% confidence interval) in model predictions which was the result of Monte Carlo simulations Given this envelope of predictive uncertainty, one can use the binomial test to examine the probability that a significant number of real observations falls outside the interval (Kirchner and Whicker 1984) In the current example, the probability that a significant number of observations falls outside the confidence interval was zero Thus, the distribution of uncertainty in model predictions covers the data distribution

Ratios of predicted to observed values can also be used to measure the model s performance against real data Assuming that the predictions and observations are lognormally distributed a paired t-test can be used to compare the ratios of validation data with simulated values (Shannon 1975) Simulated values are paired with observations by date then the mean and variance for the distribution of differences of the logarithms are computed The Student's t-test is used to test the null hypothesis that the mean of the differences is 0 i e , that the mean ratio is 1 For the example shown the geometric mean ratio of 0 92 was not statistically different from 1

Seven independent data sets have been examined to test the predictive accuracy of PATHWAY for estimating ^{131}I concentrations in milk In five of the seven sets the geometric means of predicted/observed ratios were in the 0 8-1 2 range, i e the mean PATHWAY results were within 20% of the observed data means In two cases the observed values were about 50% of predicted values Several factors including dairy cow diets an inaccurate deposition estimate etc could account for differences of a factor of two The dynamics of the model were generally in agreement with trends in the data Overall we see no reason to apply a scaling factor for the PATHWAY results for ^{131}I in milk, or a different model structure

The PATHWAY results for ^{137}Cs in milk, based on 10 data sets, were less satisfactory In general the model tended to over-predict values resulting from global fallout (and in four cases by global + Smallboy fallout) by factors of about 1 5 to 5 (Kirchner and Whicker 1984) In one case, an underprediction by 20% was noted The model dynamics corresponded, in general with the temporal

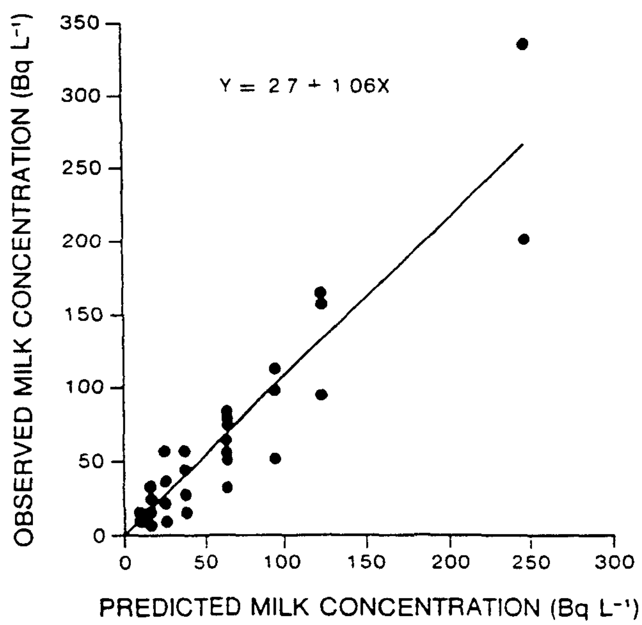

Fig 6 Observed vs predicted ^{131}I concentrations in milk for Kamas Oakley and Kimball Junction UT Data are from Fig 5

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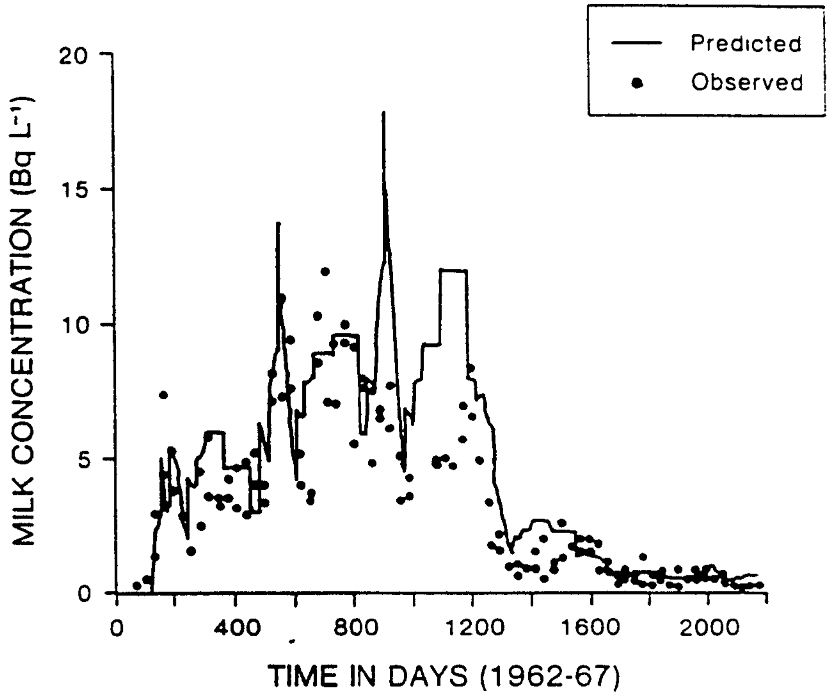

Fig 7 Predicted vs observed ^{137}Cs concentrations in milk from Salt Lake City, UT Fallout deposition measurements (Health and Safety Laboratory 1977) were used to drive the PATHWAY simulation (Kirchner and Whicker 1984)

trends in the data Example data collected by Dr R C Pendleton (deceased), Radiological Health Department, University of Utah, Salt Lake City, for ^{137}Cs in milk are shown in Fig 7 In a few cases, however, there were a significant number of runs of residuals suggesting either (1) imperfect dynamics in the model or (2) insufficient knowledge of the circumstances affecting the real data

The PATHWAY predictions for ^{90}Sr in milk did not

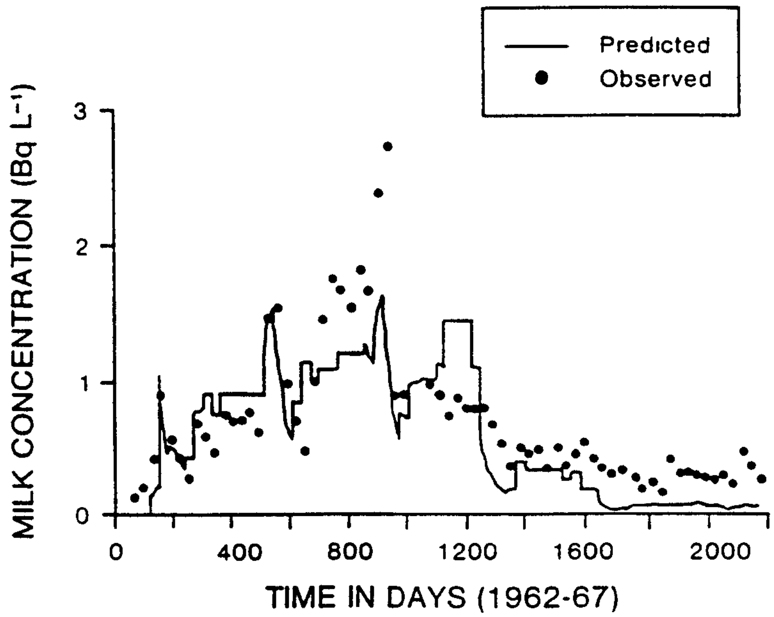

Fig. 8 Predicted vs observed ^{90}Sr concentrations in milk from Salt Lake City, UT Fallout deposition measurements (Health and Safety Laboratory 1977) were used to drive the PATHWAY simulation (Kirchner and Whicker 1984)

follow the temporal trends in the observed data from Dr R C Pendleton very well (Fig 8) In some periods PATHWAY predictions were consistently higher than observations yet in other periods the predictions were consistently low (Kirchner and Whicker 1984) However predictions were seldom different from observations by a factor of more than three In general the predictions were roughly 1 1 to 2 5 times higher than observations The temporal trends can be affected dramatically by cattle-feeding practices, including dates of hay harvests and feeding schedules These factors were not specifically known to us for the herds producing the milk that was analyzed

In the case of ^{140}Ba in milk, PATHWAY consistently (based on four data sets) underpredicted the observed data by factors of 2–4 In this case, there may be a sufficient reason to institute a scaling factor of about 3 in the PATHWAY code The dynamics of the model, however matched those of the observations

Three data sets were available on ^{137}Cs concentrations in beef In each case, PATHWAY estimates tended to be less than the observations, generally by a factor of about 2 However, the great majority of the observations fell within the 95% confidence interval of the predictions The model dynamics only corresponded with data in a general way, similar to the results for ^{137}Cs in milk

Measured concentrations of ^{137}Cs and ^{90}Sr in pasture and hay were usually within a factor of two of the model predictions Depending on the data set examined, most observations fell well within the 95% confidence interval of the predictions Particularly good agreement in values and dynamics were noted for ^{137}Cs in alfalfa hay from St George, UT Cesium-137 and ^{90}Sr concentrations in pasture from Fort Collins, CO, exceeded the predictions by an average of 25 and 50%, respectively

The model validation results indicate that PATHWAY provides reasonably accurate predictions and produces dynamic results that follow the temporal trends in real data sets Validation results for ^{131}I in milk are particularly encouraging a fortunate result since ingested ^{131}I provides the greatest dose to the thyroid gland of all pathways and milk is the primary source of ingested I The dynamics of the short-lived radionuclides apparently are easier to simulate than the temporal fluctuations of long-lived radionuclides This result is intuitive because additional pathways are involved for the long-lived radionuclides Model prediction accuracy judged by comparison to real data was always within an order of magnitude and usually within a factor of three In some cases agreement to within 20% was observed Model structure and parameter values could account for discrepancies but the lack of information on the precise circumstances of the real data sets could just as easily account for such discrepancies More effort will be required to resolve discrepancies between predictions and observations

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